

that policy makers may consider whether a change to a smaller cut-point should be considered. This is especially important in view of the possibility of a major increase in both research measurements, exposure assessment, and regulatory monitoring of fine particles, as well as of PM_{10} .

3.7.2 Size Measurements

Information on the size of fine and coarse particles comes from two basic techniques, (1) particle-counting techniques that measure the size of individual particles and convert the particle number distribution to a particle volume distribution and (2) particle-collecting techniques that use aerodynamic separation, collection of material in specific size ranges, and gravimetric or chemical analysis to determine the total mass or the mass of specific components in the size ranges collected. Particle counting has the potential advantages of not causing as much disturbance to the gas/particle equilibrium. However, considerable care must be taken to avoid heating the sample or diluting it with clean or drier air than that present in the atmosphere. With particle counting techniques it may also be possible to avoid problems of particle bounce. However, several expensive and complex instruments are required to cover the desired range of 0.001 to $100 \mu m$. Because sizes can be measured very precisely, the size ranges covered can be very small and an almost continuous function of number versus size can be obtained.

Particle collecting techniques have the advantage of obtaining size-differentiated samples for chemical analysis. The equipment used is simpler and less expensive. However, aerodynamic separation does not provide as distinct a classification by size. Large particles may bounce from their intended collection surface and be counted in smaller size ranges. Also, the requirement for long sampling times may result in averages of distributions that change with time. Particle collection techniques provide a limited number of size cuts and yield discontinuous functions of mass versus particle size.

Both techniques, however, clearly indicate the natural division of ambient air particles into fine and coarse modes with a minimum between 1.0 and 3.0 μ m diameter. Size distributions obtained with particle counting techniques tend to show a lower, broader, and more distinct minimum than distributions obtained with particle collection techniques such as impactors. The position of the minimum between the accumulation and coarse mode may vary from study to study. The peak of the fine particle mode tends to increase in size with increasing concentration

and with increasing relative humidity. Several good reviews of particle size distribution are available: physical properties of sulfur aerosols (Whitby, 1978), urban aerosols (Lippmann, 1980), trace elements (Milford and Davidson, 1985), particulate sulfate and nitrate in the atmosphere (Milford and Davidson, 1987), and coarse mode aerosol (Lundgren and Burton, 1995).

3.7.3 Appropriate Display of Size-Distribution Data

Size-distribution data, if not properly displayed, can give misleading information on the position and shape of peaks and valleys and can lead to incorrect conclusions, especially in regard to the position, width, and separation of fine and coarse modes. For this reason many workers use a histogram display obtained as follows. The mass, number, surface, or volume in each size range is divided by the difference of the logarithms of the diameters at the upper D_i and the lower D_{i-1} ends of the size range, and plotted as rectangles of width $\log D_i$ - $\log D_{i-1}$ and height, i.e. mass/ ($\log D_u$ - $\log D_e$) on a log diameter scale. This is normally shown as $\Delta C/\Delta \log D$, dM/d $\log D$, or normalized, for example, as $\Delta M/M^*\Delta \log D$. Such histogram plots are especially useful for impactor data, which normally yield fewer size intervals than particle-counting techniques. Examples of such displays are shown in Figure 3-13 (Wilson et al., 1977) and Figure 3-14a (John et al., 1990). D_{ae} is typically used when the data is presented as aerodynamic diameter and D_p when the data is presented as geometric diameter.

It is frequently desirable to draw a smooth line through the data in order to identify modes and the mass median diameters (MMD) and widths (σ_g) of modes. This can be done by fitting the data to two or more lognormal distributions, as was done in Figure 3-13 (also see Hasan and Dzubay, 1987; and Whitby-DISFIT (TSI, 1993). It is better to use an inversion process, such as originally developed by Twomey, to construct a continuous curve to represent the measurement data as shown in Figure 3-14b (John et al., 1990; Winklmayr et al., 1990). The continuous curve may then be fit to one or more log-normal distributions as shown in Figure 3-14c. However, one must be aware that log-normal distributions may not always provide good fit to actual data (see Figure 3-16). In this type of presentation the area in each rectangle or the area under a portion of a curve is proportional to the mass in

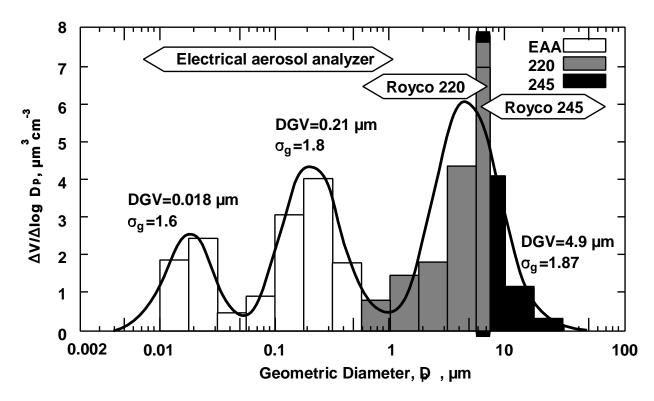


Figure 3-13. An example of histogram display and fitting to log-normal functions for particle-counting size distribution data. Instruments used and the range covered by each are shown. Counts are combined into reasonably-sized bins and displayed. Lognormal functions, fitted to the data, are shown with geometric mean diameter (DGV, equivalent to volume median diameter) of each mode and the width (σ_g) of each mode. Note the clear separation of the nuclei mode (OGV = 0.018 μ m), the accumulation mode (OGV = 0.21 μ m) and coarse mode (OGV = 4.9 μ m). Fine particles, as defined by Whitby (1978), include the nuclei and accumulation mode.

Source: Wilson et al. (1977).

that size range (or the quantity of any other parameter plotted on a linear scale). Plotting mass per impactor stage versus impactor stage number, or drawing lines connecting the midpoints of size range at the heights of the mass in each size range, does not provide such quantitative information. Once the characteristics of the impactor have been demonstrated, and once good fits to lognormal distributions have been obtained, repeated measurements of the same species may be shown by curves fitted to inversion or lognormal distributions such as the example in Figure 3-15 (John et al., 1990).

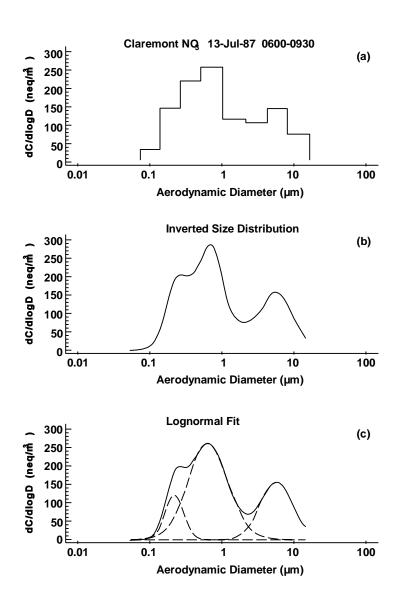


Figure 3-14. An example of an effective display of impactor data: (a) histograph showing mass found on each impactor stage and upper and lower cut points of each stage, (b) inverted size distribution, (i.e., a smooth distribution that would give the observed distribution considering the actual efficiency of each stage; cut points are not exact; each stage allows some large particles, which it should collect, to pass through to the next stage and collects some small particles which it should pass on to the next stage), (c) the solid line is the distribution obtained by fitting a sum of several lognormal functions to the inverted distribution. The dashed lines show the lognormal functions obtained from the fitting process. In this case, the use of log-normal distributions provides a reasonably good fit to the data.

Source: John et al. (1990).

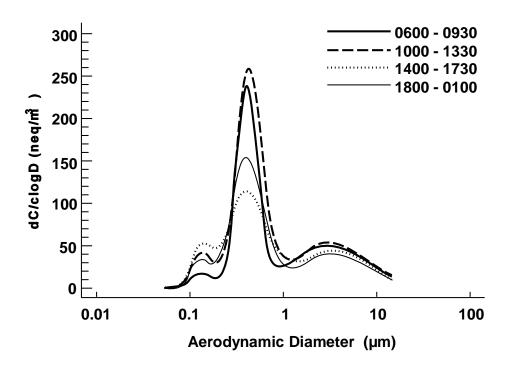


Figure 3-15. Size distributions of sulfate, Long Beach, June, 1987, showing use of fitted log-normal distributions to describe diurnal variations in size and concentration.

Source: John et al. (1990).

In impactor measurements, the maximum size of the upper stage and the minimum size of the lower stage (or after filter) are not well defined. Therefore, an arbitrary choice must be made in order to define the Δ log D_{ae} . This choice can have a remarkable influence on the perceptions of the positions, height, and width of modes. A particularly dramatic example is shown in Figure 3-16, from Šega and Fugaš (1984). The authors chose 0.1 μ m for the lower limit and 20 μ m for the upper limit, suggesting a bimodal distribution with a fine mode MMAD at about 1.5 μ m, and a coarse mode MMAD at about 10.5 μ m. However, if 0.4 μ m is chosen for the lower limit and 10 μ m for the upper limit, the display suggests a fine mode MMAD of about 0.7 μ m and a coarse mode MMAD of about 8 μ m.

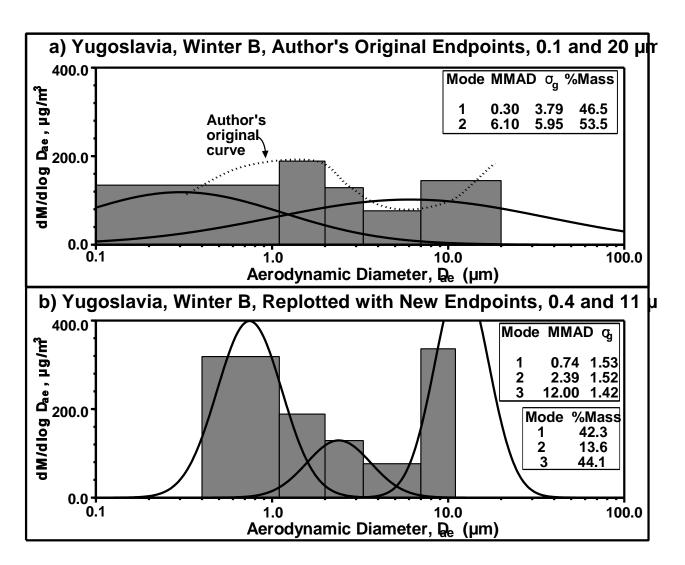


Figure 3-16. Effect of changing endpoints. This example of impactor data shows how the lack of a well-defined upper and lower size limit can affect the perception of the location of fine and coarse particle modes. A histogram with an upper limit of 20 μ m and a lower limit of 0.1 μ m diameter, along with the curve drawn by authors of the report, is shown in Figure 3-16a. In Figure 3-16b a histogram with a lower limit of 0.4 μ m and an upper limit of 10 μ m is shown. The author's free hand curve suggests a fine particle MMAD around 1.5 μ m diameter. A quite different idea of the location of the modes is given when different endpoints are chosen. Much of the material found between 1.0 and 5.0 μ m is probably smaller particles caught on the glass fiber impactor stages which have very poor separation efficiencies. The data has been fitted to a 3-lognormal mode distribution; however, log-normal distributions do not provide a good fit to this data.

Source: Šega and Fugaš (1984).

3.7.4 Comparison of Particle-Counting and Particle-Collection Techniques

Unfortunately, there have been few efforts to compare results of the two particle-sizing techniques. One such effort is shown in Figure 3-17 (Durham et al., 1975). The differences between the two techniques, as evident in the figure, are qualitatively observed in individual studies using either of the two techniques. Particle counting techniques usually give a lower and wider minimum. Typically particle counting leads to volume distributions plotted versus geometric size (or more properly, geometric size inferred from mobility or optical size); whereas impactor separations give mass versus aerodynamic size. In Figure 3-17 both geometric and aerodynamic scales are given. This figure illustrates the problems involved in defining particle "size" and serves as a reminder that each particle sizing technique gives a different "size". The upper scale, used for impactor data, is given in aerodynamic diameter. The aerodynamic diameter of a particle is the diameter of a particle of density, $\rho=1.0$, which would behave similarly with respect to impaction as the particle in question. For spheres in the coarse mode, the aerodynamic diameter, D_a , equals $\sqrt{\rho} D_p$, where ρ is the density of the particle and D_p is the geometric diameter. Since coarse particles are expected to have a greater density than fine particles, converting the volume, geometric-size distribution to a mass, aerodynamic-size distribution would increase the apparent size of the volume distribution above 1 μ m and widen the minimum. For small particles, below 0.5 μ m, or at reduced pressures where the mean free path of the gas molecules is of the same order, or larger than the particle diameter, the Stokes diameter, which is more closely related to the diffusion coefficient, is a more useful parameter. The relationships between Stokes, aerodynamic and geometric diameter are discussed in Section 3.1.3.1.

The particle size distribution shown on the bottom of the graph was derived from a combination of a mobility counter and several optical counters. The "mobility size", obtained from the electrical aerosol analyzer (EAA) in earlier studies and the differential mobility analyzer (DMA) in more recent studies is dependent on the particle shape but not the density. For irregularly shaped particles the "mobility" size gives the Stokes diameter, which is the geometric diameter of a sphere with the same aerodynamic drag. For a sphere the Stokes diameter and the geometric diameter are the same. By comparing the mobility or Stokes diameter to the aerodynamic diameter it is possible to measure the density of spherical particles (Stein et al., 1994).

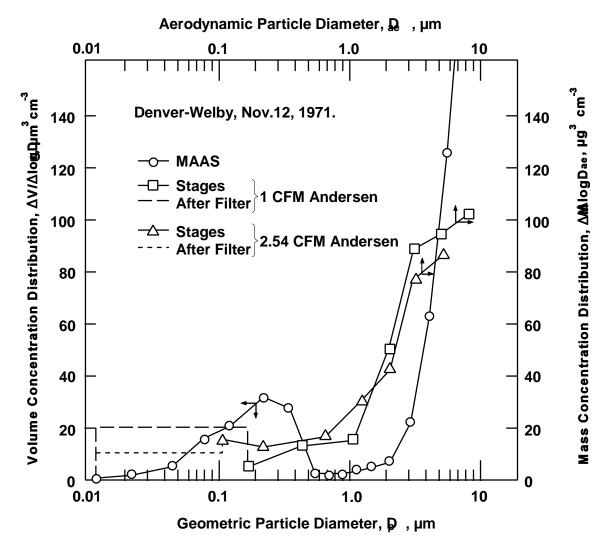


Figure 3-17. These size distributions, obtained during an EPA study of the Denver brown cloud represent one of the few efforts to compare particle-counting and particle-collection size-distribution measurements. Note that impactor data is given in aerodynamic diameter, $D_{\rm ae}$, and particle-counting data is given in geometric diameter, $D_{\rm p}$, derived from the number distribution and estimated density.

Source: Durham et al. (1975).

The "optical" size of a particle depends on the particles shape and refractive index, and on the characteristics of the optical counter. The amount of light scattered by a particle at a wavelength near the particle size varies rapidly with changes in size, wavelength, refractive index, and scattering angle. Therefore, several different optical counters may be needed to cover the range of atmospheric particle sizes. Because of non-linearities in the response of laser or

narrow wavelength optical counters to size changes it is especially difficult to measure particles in the 0.5 to 1.0 size range (Hering and McMurry, 1991; Kim, 1995). Since the amount of scattered light depends strongly on the refractive index it would be useful to calibrate optical counters with particles of the same refractive index as those in the atmosphere. Hering and McMurry (1991) used a differential mobility analyzer to select particles of a uniform geometric diameter. The light scattering of these monodispersed atmospheric particles, as measured by a Particle Measuring System LAS-X optical counter, was compared to that of spheres of polystyrene latex (a substance frequently used to calibrate optical counters) and oleic acid of the same geometric diameter. The atmospheric aerosols scattered less light than polystyrene latex sphere (refractive index = 1.9 - 0.0i), but about the same amount of light as oleic acid spheres (refractive index = 1.46 - 0.0i) of the same geometric size. Relating the variety of sizes measured by particle counters and impactors, and displaying them together on an aerodynamic diameter scale, or other scale, is a major task which has not yet been adequately addressed.

The greater width of the coarse modes, as measured by the impactor in Figure 3-17, may be attributed to the use of glass fiber filter paper for the impactor surface. It is now recognized that the use of glass fiber filter material, as contrasted to a flat surface, causes a severe reduction in the effectiveness of the cut. Large particles bounce off the glass fiber (Vanderpool et al., 1987) giving much reduced collection efficiencies; whereas fine particles penetrate into the fiber and some are captured in stages that should have near zero collection efficiencies (Rao and Whitby, 1978). Many studies that used the Anderson High Volume Fractionating Sampler also used glass fiber filters. The use of glass fiber filters as impaction collection surfaces causes any given size range to contain both larger and smaller particles than predicted and thus tends to spread out the modes and fill in the minima. An example of the smoothing effect of glass fiber collection surfaces, and especially the collection of fine particles on upper stages, can be seen in Figure 3-16. Nevertheless, the bimodal nature of the ambient aerosol is still captured.

3.7.5 Review of Size-Distribution Data

3.7.5.1 Early Studies

In 1978, when EPA scientists debated the best cut-point to separate fine particles from coarse particles, there was limited information available. Particle-counting data from California studies had been summarized by Whitby and Sverdrup (1980) and are shown in Figure 3-18. With the exception of one distribution from Pomona, all distributions showed a minimum near 1 μ m and indications of significant amounts of coarse particle material between 1.0 and 2.5 μ m. (The region between 1 and 2.5 μ m will be referred to as the intermodal region.) Other studies of size distribution (McMurry et al., 1981) in the Southeastern United States, provided similar information (Figure 3-19).

Results from several impactor studies were also available, some of which suggested two modes. However, much of the impactor data were considered unreliable in regard to the existence and position of modes (Whitby et al., 1974). However, one of the more extensive and reliable studies available (Patterson and Wagman, 1977) provided confirmation of the Whitby bimodal observations. In this study, mass and composition measurements were made for four different levels of visibility. The histograms for mass, sulfate, and iron for two levels of visibility are shown in Figure 3-20. It is clear that the major portion of the fine mass is below 0.6 μ m and the major portion of the coarse mass is greater than 3 μ m in diameter. These impactor data, coupled with the more extensive number-size distributions data of Whitby and Sverdrup (1980) led to a preference for a 1 μ m cut-point but an acceptance of 2.5 μ m on the assumption, then considered to be the case, that 2.5 μ m represented the minimum cut-point that was attainable with a dichotomous sampler (Miller et al., 1979).

3.7.5.2 Recent Work

In the intervening 15 years, there has been very little additional work in which particle-counting techniques, covering the entire size range, have been used to measure ambient aerosols. Most of the particle-counting studies have focused on fine and ultrafine particles, diameter <1.0 μ m. There have however been a number of impactor studies that provide insight into the size of the fine and coarse modes and into what material is found between them.

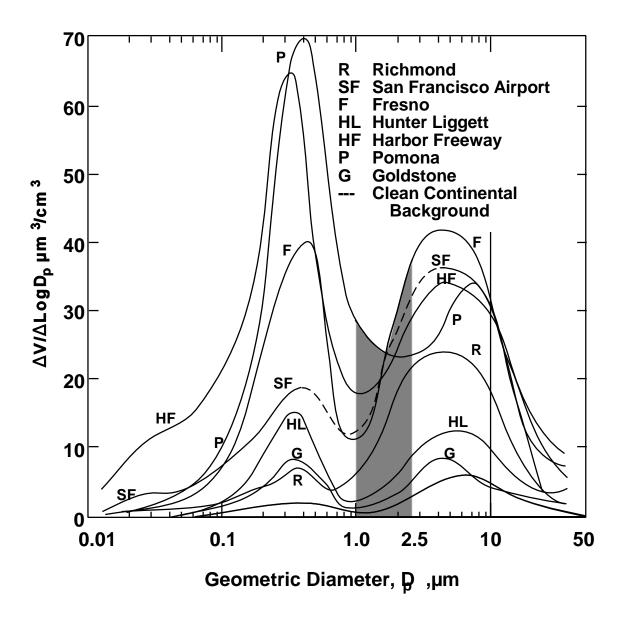


Figure 3-18. Grand average volume-size distributions from the Aerosol Characterization Experiment (ACHEX) in 1972. A size distribution for clean continental aerosol is shown for comparison. Note that with the exception of the Pomona size distribution, all distributions show a distinct minima near 1.0 μ m diameter. A line has been added at 1.0 μ m, 2.5 μ m, and 10 μ m diameter to indicate how much of the coarse particle mode is observed between 1.0 and 2.5 μ m diameter.

Source: Whitby and Sverdrup (1980).

There are only a few impactor size distribution studies that cover the full size range from 0.01 to 100 μ m (Lundgren and Hausknecht, 1982a,b; Lundgren et al., 1984; Burton and Lundgren, 1987; Vanderpool et al., 1987). Lundgren and co-workers used a mobile

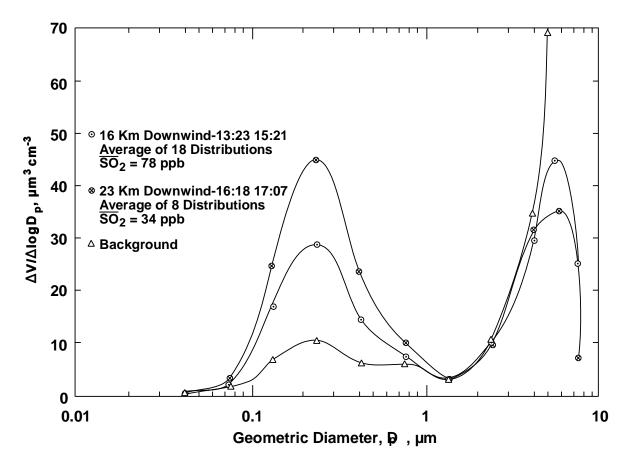


Figure 3-19. Volume-size distribution taken in the midwestern United States near the Cumberland Power Plant in Tennessee. Note that coarse mode decreases and fine mode increased as the mobile sampling van moved downwind farther from urban influence but allowing more time for reaction as the power plant plume mixed with background air and SO_2 was converted to sulfate and SO_3 to nitrate.

Source: McMurry et al. (1981).

unit, the wide range aerosol classifier (WRAC), to measure mass-size distribution in ten size ranges from <0.4 to >50 μ m. Two distributions, averages for Philadelphia and Phoenix, are shown in Figure 3-21. Both clearly indicate a fine particle mode with an MMAD near 0.5 μ m for Philadelphia and below 0.3 μ m for Phoenix. Both show a coarse particle mode with an MMAD near 20 μ m in diameter. However, there is a significant amount of material found in the intermodal region, 1 to 2.5 μ m. Although the intermodal mass is not a significant fraction of the total suspended particulate mass or even of TSP, as would be measured by a high-volume sample (upper cut-point around 25 μ m), it does represent a

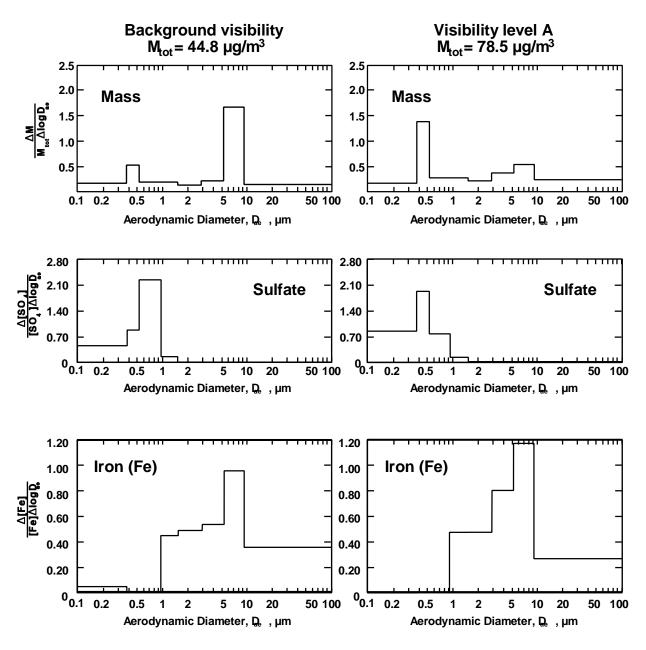


Figure 3-20. Examples of size distribution histograms for total mass, sulfate, and iron obtained at two visibility levels using an Andersen impactor. Arbitrary choice of 0.1 and 100 for lower and upper limits cause the extreme rectangles to be long and low. Note the separation into fine and coarse modes in mass and that sulfate and iron clearly belong in the fine and coarse mode respectively.

Source: Patterson and Wagman (1977).

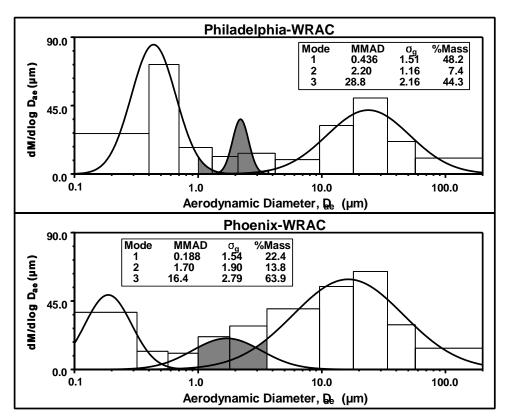


Figure 3-21. Impactor size distribution measurement generated by Lundgren et al. with the Wide Range Aerosol Classifier: (a) Philadelphia and (b) Phoenix. Note the presence of more coarse mode particles in the size range 1 to 2.5 μ m, in the dryer environment of Phoenix.

Source: Adapted from Lundgren and Hausknecht, 1982b.

major portion of the coarse fraction of PM_{10} . An attempt has been made to fit the distribution with three, log-normal distributions. In this case, the fit is poor. In the Phoenix case the accumulation mode cannot be defined other than that the MMAD is below 0.2 μ m. The coarse particle fractions are very wide suggesting the possibility of two or more modes (Figure 3-24). The material between 1 and 2.5 μ m is not a new mode but an indication of either an artifact due to particle bounce, or a long-lasting tail of the coarse distribution.

The existing size-distribution data were recently reviewed by Lundgren and Burton (1995), with emphasis on the coarse mode. They concluded that the coarse mode could be reasonably well described by a lognormal distribution with a mass median aerodynamic diameter (MMAD) of 15 to 25 μ m and a mode spread (σ_g) of approximately two. This allows one to calculate, for a freshly-generated coarse mode aerosol, that about 1% of the

mass would be less than 2.5 μ m and only about 0.1% would be less than 1.0 μ m in diameter. This conclusion is confirmed by data from Whitby in which a wind change allowed a measurement of fresh coarse mode aerosol (National Research Council, 1979). As can be seen in Figure 3-22, the intermodal mass, 1.0 to 2.5 μ m, was not affected, even though the mass at 20 μ m increased substantially.

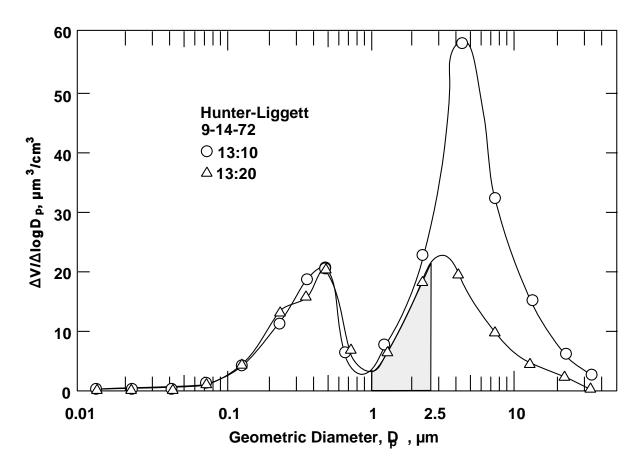


Figure 3-22. Example of aged and fresh coarse mode particle size distributions. A sudden wind change brought fresh wind-blown dust to the sampler, operated as part of the South Coast Air Quality Study. Note that there is only a very small change in the intermodal mass, 1.0 to 2.5 μ m diameter, although there is a major increase in the mass between 2.5 and 10 μ m in diameter.

Source: National Research Council (1979).

Another extensive set of studies covering the full size range, but limited to the Chicago area, has been reported by Noll and coworkers (Lin et al., 1993, 1994). They used an Andersen impactor for smaller particles and a Noll Rotary Impactor for larger particles. Results of Lin et al. also indicate a bimodal mass distribution. For the shorter time interval measurements (8 or 16 h), the average MMAD for the fine mode was $0.42~\mu m$, with a σ_g around two. The average MMAD of the coarse mode was $26\pm 8~\mu m$, with a σ_g varying from 2.0 to 3.5. As shown in Figure 3-23, the results of Noll and coworkers (Lin et al., 1993, 1994) also suggest that in some instances little coarse mode material is found in the intermodal region, 1.0 to $2.5~\mu m$. Lin et al. (1993) combined material on the 0.65 to $1.0~\mu m$ and the 1.0 to $2.0~\mu m$ stages before weighing. Therefore, the MMAD of the accumulation mode is not as well defined as it might be, and could be smaller than that given by the fitting process. Therefore, these results cannot be used to show that some fine PM is found above $1.0~\mu m$. When fitted to two log-normal distributions the fit is poor and the coarse mode is very wide. The fit with three log-normal distributions is used to show the possibility of particle bounce or a second mode within the coarse particle size range contributing to mass in the intermodal (1-2.5 $~\mu m$) region.

3.7.6 Intermodal Region

3.7.6.1 Coarse Mode

The question then arises, what portion of the coarse mode material found in the intermodal region is real and what portion is artifact? As discussed in Section 3.3.3.2.4, the optical size may differ from the geometric or aerodynamic size. Optical counters are normally calibrated with latex particles, or other particles of a specific refractive index. Atmospheric particles with different refractive indices would be incorrectly sized if the difference in refractive index resulted in a difference in the amount of light scattered by the particles (Wilson et al., 1988; Liu et al., 1992; Hering and McMurry, 1991). For particle counters using lasers, particles of different sizes within the 0.5 to 1.0 μ m range may give the same light scattering (Hering and McMurry, 1991; Kim 1995).

In the case of impactors, it is possible that an artifact may arise from particle bounce, from fragmentation of larger agglomerates, or from loosening of material from other surfaces by impacting particles. The problem of particle bounce in impactors has been treated

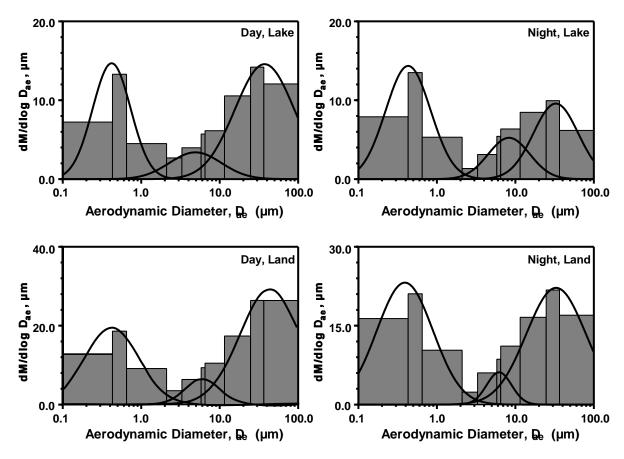


Figure 3-23. Size distributions reported by Noll and co-workers from the Chicago area using an Anderson impactor for the smaller particles and a Noll Rotary Impactor for the larger particles.

Source: Lin et al. (1993).

theoretically and practically in many studies (Wang and John, 1987, 1988). Most workers coat the coarse particle stages with a grease or oil to reduce bounce. However, as the surface becomes covered with aerosols, a particle may impact another particle instead of the surface and either bounce to a lower stage or cause deagglomeration and reentrainment of previously collected particles (John et al., 1991; John and Sethi, 1993). As impactor collection plates become loaded or as inlet upper size cut surfaces become dirty, the magnitude of the effect increases (Ranade et al., 1990; John and Wang, 1991). One result is a lowering of the effective cut point of the inlet and the impactor stages. Thus, it is uncertain how much of the mass found in the intermodal size range is real and how much is due to artifacts.

There are several reasons to believe, however, that some of the intermodal mass may be real. For example, Lundgren and Burton (1995) point out that the lifetime of particles in the atmosphere is a strong function of their aerodynamic size. Thus, while freshly generated coarse mode aerosol may have a MMAD of 20 μ m, with only 1% below 2.5 μ m, as the aerosol ages the larger particles will rapidly fall out, leaving a distribution enriched with particles in the small-size tail of the distribution.

A second explanation has to do with the possible multimodal nature of dust generated by wind or vehicular traffic. A study by the U.S. Army (Pinnick et al., 1985) measured the size distribution of dust generated by heavy vehicles driven on unpaved roadways in the arid southwestern United States. A variety of light-scattering instruments were used and were recalibrated for the refractive index of the soil particles. The occurrence of strong surface winds (gusts of 15 to 20 m s⁻¹) during the study permitted, in addition to the vehicular-generated dust, some measurements of windblown dust. There were some differences between sandy soil and silty soil, and between dust generated by vehicular traffic and by wind. However, all situations produced a bimodal size distributions. The upper mode had an MMAD that ranged from 35 to 53 μ m, with σ_g from 1.37 to 1.68. Of particular interest, however, was a second mode having an MMAD that varied from 6.2 to 9.6 μ m, with a σ_{g} from 1.95 to 2.20. (One measurement in silty soil had an MMAD of 19.4 μ m.) The MMADs of the smaller coarse particle modes are significantly smaller than those coarse mode MMADs observed by Lundgren or Noll. An example of vehicular generated dust is shown in Figure 3-24. Note that the differential mass is plotted on a logarithmic scale. These results suggest that in arid areas, significant soil material, generated by traffic or wind, may be found in the intermodal region.

A third reason for believing that the intermodal mass is real has to do with the relative size efficiency of particle removal equipment used on power plants and other large industrial facilities. Older control devices, which may still be used in some applications, allow significant particle mass to escape. Overall mass efficiencies are approximately 80% for cyclones and 94% for scrubbers. Modern control devices have very high overall efficiencies, 99.2% for electrostatic precipitators (ESP) and 99.8% for baghouses. However, all of these devices have efficiencies for coarse particles that decrease with decreasing size. Efficiencies typically reach a minimum between 0.1 and 1 μ m and increase for particles

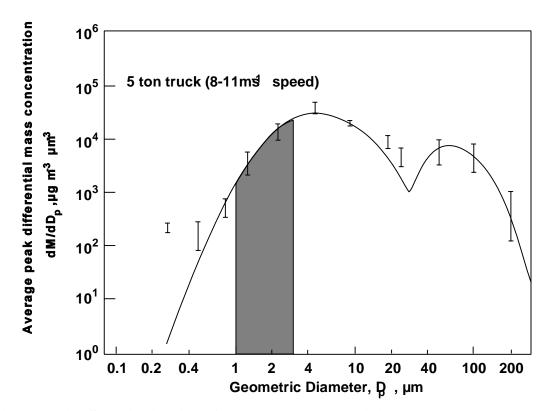


Figure 3-24. Size distribution of dust generated by driving a truck over an unpaved test track. "Error bars" show the range of distributions from individual tests. The curves shown are log-normal fits to the average distribution. The original data were plotted as log radius but have been replotted versus log diameter. The shaded bar between lines at diameters of 1.0 and 2.5 μ m indicates how the smaller size mode of this dust can contribute to the intermodal mass found in arid areas (see Figures 3-21 and 3-23).

Source: Pinnick et al. (1985).

smaller than $0.1~\mu m$. Thus, although most of the particulate mass is captured, the particles that do escape are in the smaller size range. Data from U.S. EPA, plotted in Figure 3-25, (U.S. Environmental Protection Agency, 1995) show the size distribution of fly ash from a pulverized coal power plant and the size distribution of the material escaping from the various control devices. The small-size tail of the coarse mode escapes preferentially and may possibly contribute material to the intermodal region.

Cheng et al. (1985) reported experimental measurements from an atmospheric fluidizedbed coal combustor. Size distribution measurements, made downstream of a cyclone and again downstream from baghouse filtration of the material leaving the cyclone, are shown in

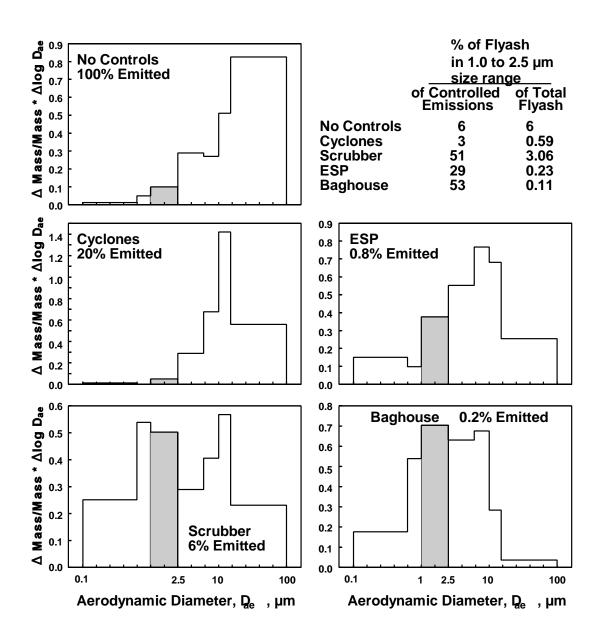


Figure 3-25. Size distribution of emissions from a pulverized-coal power plant and the particle size distributions remaining after several types of control devices.

Source: U.S. Environmental Protection Agency (1995).

Figure 3-26 (Cheng et al., 1985). Electron microscope photographs confirmed a fine particle mode of spherical particles between 0.1 and 0.25 μ m, presumably formed from evaporation and condensation of volatile species from the coal matrix; and irregular-shaped chunks from the coarse mode with a peak concentration between 1 and 3 μ m in diameter.

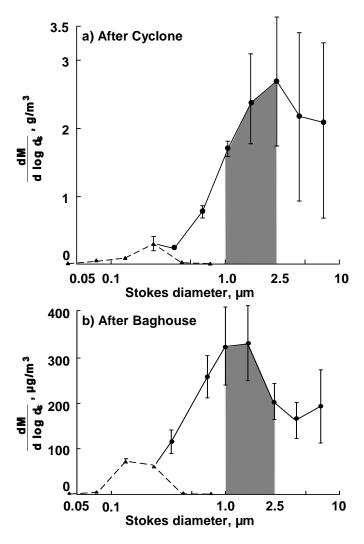


Figure 3-26. Size distributions from a fluidized-bed, pulverized coal combustor, (a) after initial cleanup by a cyclone collector and (b) after final cleanup by a baghouse.

Source: Cheng et al. (1985).

A fourth piece of evidence comes from studies in which measurements are made of the elemental composition of $PM_{2.5}$ and PM_{10} or the coarse fraction of PM_{10} . Elements representative of soil type material have been found in the $PM_{2.5}$ fraction. In a study in Philadelphia that used dichotomous samplers, an amount of soil-type material equal to 5% of the coarse mode fraction of PM_{10} was found in the $PM_{2.5}$ fraction (Dzubay et al., 1988). Since the virtual impactor used in the dichotomous sampler minimizes particle bounce and reintrainment, this would appear to be the small-size tail of the coarse mode in the 1 to 2.5 μ m size range.

Similar results have been reported from the IMPROVE network (Eldred et al., 1994). Elemental analysis suggested that soil-derived material, equal to 20% of the coarse fraction of the PM_{10} sample, was found in the PM_{25} sample.

Thus, one can conclude that coarse mode material is found in the intermodal region. There are reasons to suspect that a portion of this material is an artifact but that a portion is real coarse mode material having an aerodynamic diameter between 1.0 and 2.5 µm. In either event, this can lead to a misunderstanding of the source of the particles, to inappropriate control strategies, or to exposure studies that fail to differentiate correctly between fine and coarse particles.

3.7.6.2 Fine Mode

This section discusses the source of fine mode material found in the intermodal region. Early particle-counting data suggested that, with a few exceptions, significant mass of fine mode material would not be found above 1 μ m (see Figures 3-13, 3-18, 3-19, and 3-20). However, impactor studies, on some occasions, have observed significant mass on stages with a cut point of 1 μ m. In some instances, the MMAD of the fine mode was as large as 1 μ m (John et al., 1990). The change in relative humidity produced by a few degrees change in temperature can significantly modify the MMAD of an ambient aerosol size distribution. As the RH approaches 100%, accumulation mode aerosols, with dry sizes below 1.0 μ m in diameter, may grow larger than 2.5 μ m in diameter, be rejected by PM_{2.5} samples, and be counted as coarse particles.

Before examining additional field data demonstrating the effect of relative humidity on particle size, it is useful to review some basic information on the interaction of water vapor with the components of fine particles. Sulfuric acid (H_2SO_4) is a hygroscopic substance. When exposed to water vapor a H_2SO_4 droplet will absorb water vapor and grow in size until an equilibrium exists between the liquid water concentration in the H_2SO_4 solution droplet and the water vapor concentration in the air. The amount of water in the sulfuric acid droplet will increase and decrease smoothly as the RH increases and decreases. Ammonium sulfate, $(NH_4)_2SO_4$, however, is deliquescent. If initially a crystal in dry air, it will remain a crystal until a certain RH is reached; at this point it will absorb water and become a solution droplet. The RH at which this happens, $\approx 80\%$ RH in the case of

NH₄)₂SO₄, is called the deliquescent point. At RH's above the deliquescent point the (NH₄)₂SO₄ droplets are hygroscopic, gaining or losing water reversibly as the RH increases or decreases. If the RH decreases below the deliquescent point the solution droplet becomes supersaturated and unstable to crystallization. However, sub-micron sized droplets will remain supersaturated until a significantly lower RH, known as the crystallization or efflorescent point is reached. The crystallization point decreases with decreasing droplet size and decreasing purity (Whitby, 1984). Thus, for a deliquescent substance, a plot of droplet diameter or water content as a function of RH will have two lines, one for increasing RH and another for decreasing RH. An example of this phenomenon, known as hysteresis, is shown in Figure 3-27. Table 3-16 shows the RH at the deliquescent and crystallization points for some compounds found in the atmosphere.

Much experimental and theoretical effort has gone into understanding this process. The basic theory was elucidated by Hänel (1976). Much experimental work has been done on atmospheric species (e.g., Tang and Munkelwitz, 1977, 1993; Richardson and Spann, 1984). The electrodynamic balance, by which single particles can be studied, has advanced the understanding of particle-water vapor equilibrium, especially for particles in metastable states, e.g., the supersaturated solution particles which are responsible for the hysteresis loop shown in Figure 3-27 (Cohen et al., 1987a,b; Chan et al., 1992; Kim et al., 1994). Ammonium nitrate, NH₄NO₃, because of its volatility, is difficult to handle but has been studied successfully by Richardson and Hightower (1987). The aerosol equilibria models developed by Seinfeld and coworkers allow calculation of the water content of bulk solution as a function of relative humidity (Kim and Seinfeld). The model SCAPE (Kim et al., 1993a,b) has been used to estimate the contribution of water to suspended aerosol mass in the California South Coast Air Basin using particle composition data from the 1987 Southern California Air Quality Study (Meng et al., 1995). From midnight to early morning, when the temperature is low and relative humidity is high, water was usually the predominant aerosol substance. Particulate water in the winter was estimated to be considerably larger than in the summer at each of the four sites studied.

The water content of a sub-micron sized droplet, and therefore its size, depends not only on the dry size but is a result of a balance between surface tension and solute concentration (Li et al., 1992). Pure water is in equilibrium with its vapor when the RH

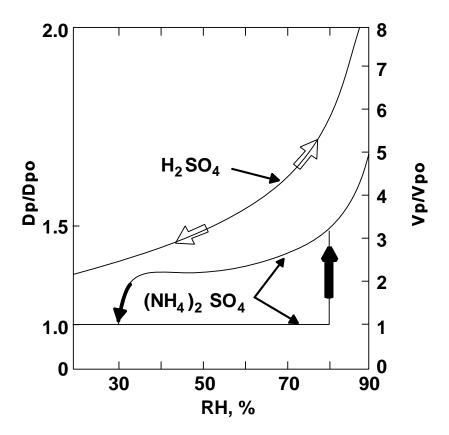


Figure 3-27. Particle growth curves showing fully reversible hygroscopic growth of sulfuric acid (H_2SO_4) particles, deliquescent growth of ammonium sulfate $[(NH_4)_2\ SO_4]$ particles at about 80% relative humidity (RH), hygroscopic growth of ammonium sulfate solution droplets at RH greater than 80%, and hysteresis (the droplet remains supersaturated as the RH decreases below 80%) until the crystallization point is reached.

Source: National Research Council (1993) adapted from Tang (1980).

TABLE 3-16. RELATIVE HUMIDITY OF DELIQUESCENCE AND CRYSTALLIZATION FOR SEVERAL ATMOSPHERIC SALTS^a

Compound	Deliquescence	Crystallization ^c
$(NH_4)_2SO_4$	79.9 ± 0.5	37 ± 2
NH ₄ HSO ₄	39.0 ± 0.5^b	
NH_4NO_3	61.8	
NaCl	75.3 ± 0.1	42

^aTaken from Tang and Munkelwitz (1993) unless otherwise indicated.

^bTang and Munkelwitz (1977).

^cShaw and Rood (1990) and references therein.

equals 100% and is therefore, stable, i.e. the rate of evaporation equals the rate of condensation. The water in a solution will be in equilibrium with water vapor at a lower water vapor concentration because the presence of solute molecules or ions lower the rate of evaporation. Therefore, a solution will absorb water and become more dilute, increasing the water vapor concentration needed for equilibrium until the solution water vapor concentration required for equilibrium matches the ambient water vapor concentration or RH. As the droplet size decreases the surface tension increases and the vapor pressure of water required to maintain equilibrium increases. Therefore, the smaller the dry size of the particle, the less the amount of growth as RH increases.

Theoretical calculations of the growth of various sizes of ammonium sulfate particles and an experimental verification of such calculations, using a simulation of the humidification process in the human lung, are shown in Figure 3-28. Note the very rapid increase in the amount of water and in the diameter of the aerosol particle as the relative humidity approaches 100% RH. Considering the difficulty of measuring relative humidity accurately between 99 and 100%, theory and experiment are in reasonable agreement. As can be seen the effect of surface tension is most important for particles with dry size less than 100 nm (0.1 μ m). This phenomenon may be of importance in considering the biological effect of water-soluble pollutants. Accumulation mode particles will be diluted when exposed to humidification in the lungs. Ultrafine or nuclei mode particles will not be diluted as much. In the atmospheric aerosol the number distribution will almost always be dominated by particles below 100 nm (see Section 3.1.2). However, aerosols generated in the laboratory for exposure studies probably lack the smaller particles found in the atmosphere. This provides a hypothesis for the difference in effects observed in epidemiological studies and laboratory exposure studies. The importance of this more concentrated, ultrafine droplet component of the atmospheric aerosol may have been neglected because most of the experimental studies of hygroscopicity have used near-micron-sized particles. However, in the modeling of deposition of hygroscopic particles, workers, such as Martonen (1993), have corrected the experimental curves of particle size as a function of RH, based on measurements of near micron-sized particles, to account for the effects of surface tension on ultrafine particles.

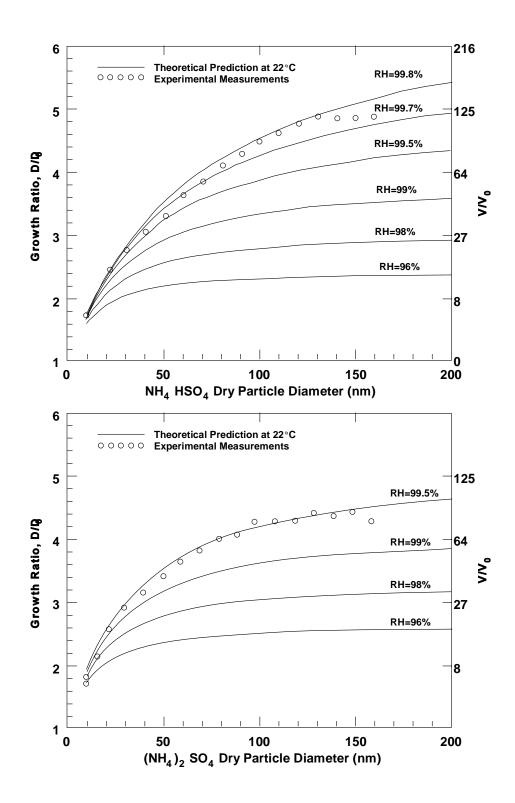


Figure 3-28. Theoretical predictions and experimental measurements of growth of NH_4HSO_4 and $(NH_4)_2SO_4$ particles at relative humidity between 95 and 100%.

Source: Li et al. (1992).

In addition to the laboratory studies discussed above there are some measurements on the effect of RH changes on atmospheric aerosol. McMurry and co-workers have made use of a Tandem Differential Mobility Analyzer (TDMA) system (Rader and McMurry, 1986) to measure the change in particle size with changes in relative humidity at Claremont, CA, as part of the Southern California Air Quality Study (SCAQS) (McMurry and Stolzenberg, 1989) and at the Grand Canyon National Park, AZ, as part of the Navajo Generating Station Visibility Study (Zhang et al., 1993; Pitchford and McMurry, 1994). One mobility analyzer is used to isolate a narrow size distribution. After humidification the size distribution of this fraction is measured. An example is shown in Figure 3-29. Note that Figure 3-29 is a number size distribution not a mass size distribution. Particle growth with increasing RH is evident. However, between 70 and 91% RH the distribution splits into less-hygroscopic and more-hygroscopic components. Pitchford and McMurry (1994) attribute this splitting to external mixing, i.e. there are two relatively distinct classes of particles, both containing some soluble and some non-soluble material, with the more hygroscopic component containing significantly more soluble and hygroscopic material. A summary of the results of these studies is given in Table 3-17 (Zhang et al., 1993). The difference in growth rates may be due both to size and to variation in composition as a function of size. The lower growth factor for $0.2 \mu m$ particles in Claremont relative to the Grand Canyon may be due to a higher concentration of non-soluble organic material in Claremont.

While there is a significant amount of information on the hygroscopic properties of inorganic compounds, much less is known about the hygroscopic properties of organic components of the atmospheric aerosol. Saxena et al. (1995) have examined the hygroscopic properties of several organic species and noted that water soluble organics may be hygroscopic or deliquescent. Using concurrent cascade impactor samples, they determined the composition of the Grand Canyon and Claremont aerosol, whose size distribution as a function of relative humidity was discussed above. They compared the observed water content at the higher relative humidity with the water content calculated for the inorganic components. They concluded "that the aggregate hygroscopic properties of inorganic particles are altered when organics are also present. Furthermore, the alterations can be positive or negative. The findings are consistent with the expectation that organics are

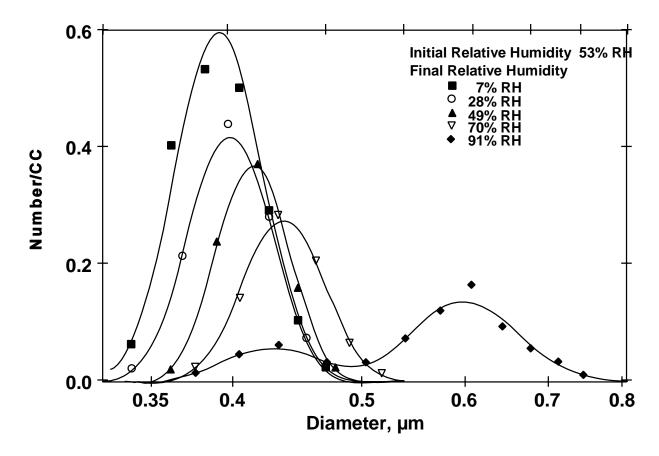


Figure 3-29. Tandem Differential Mobility Analyzer measurements of the sensitivity of particle size to relative humidity at Claremont, CA. Particle number concentrations varied during the measurement, therefore changes in relative size with humidity are meaningful but changes in number concentration are not.

Source: McMurry and Stolzenberg (1989).

predominantly secondary (and thus likely to be hydrophilic) in nonurban areas and predominantly primary (and hence hydrophobic) in urban areas".

Some experimental examples of the significant effect of relative humidity on ambient aerosol size distributions are shown in Figure 3-30 (Lowenthal et al., 1995). In this work, impactor collection and ion chromatographic analysis were used to measure sulfate size distributions over short enough periods to demonstrate the effects of changing relative humidities. The results suggest that the lognormal distribution is preserved as relative humidity increases, but that the MMAD increases. This effect is especially pronounced as the relative humidity approaches 100%.

TABLE 3-17. SUMMARY OF HYGROSCOPIC GROWTH FACTORS^a

1987 SCAQS, Claremont, CA

Dry Size (μm)	More Hygroscopic Peak $\frac{D_{p}(90 \pm 3\% \text{ RH})}{D_{p}(0\% \text{ RH})}$	Less Hygroscopic Peak $\frac{D_{p}(87 \pm 2\% \text{ RH})}{D_{p}(0\% \text{ RH})}$
0.05	1.14 ± 0.05	1.03 ± 0.03
0.2	1.23 ± 0.08	1.02 ± 0.02
0.4	1.63 ± 0.11	1.04 ± 0.05
0.5	1.59 ± 0.08	1.07 ± 0.03

1990 NGS Visibility Study, Grand Canyon, AZ

	More Hygroscopic Peak $D_0(89 \pm 4\% \text{ RH})$	Less Hygroscopic Peak $\underline{D}_{n}(89 \pm 4\% \text{ RH})$
Dry Size (μm)	D _p (0% RH)	$D_{p}(0\% \text{ RH})$
0.05	1.36 ± 0.08	1.14 ± 0.10
0.10	1.42 ± 0.08	1.17 ± 0.09
0.20	1.49 ± 0.11	1.17 ± 0.10
0.30	1.51 ± 0.09	1.14 ± 0.10
0.40	1.43 ± 0.10	1.07 ± 0.03

^aValues are mean ± standard deviations.

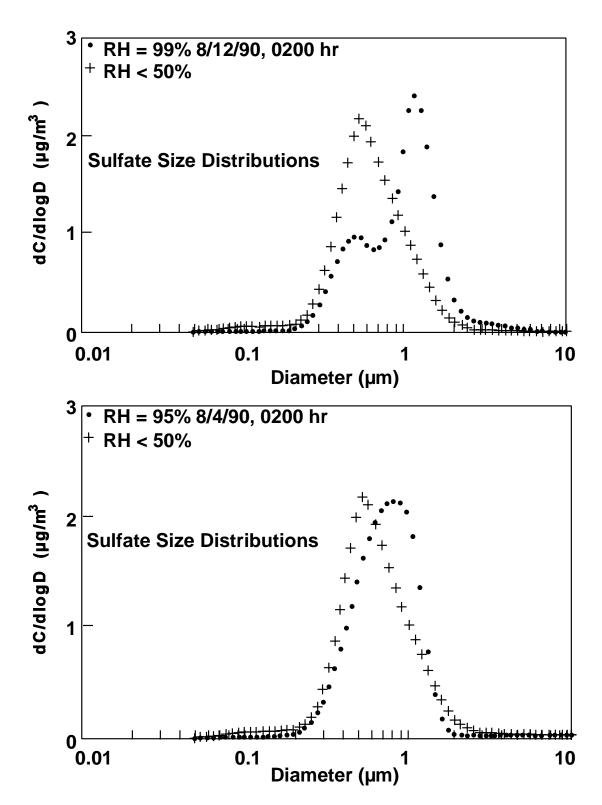


Figure 3-30. Example of growth in particle size due primarily to increases in relative humidity from Uniontown, PA.

Source: Lowenthal et al. (1995).

There are also studies of the behavior of ambient aerosols as the relative humidity is reduced by heating the sampled air. Shaw and Rood (1990) report a study using a heated integrating nephelometer in which crystallization RHs of 4 to 67% were observed. Similar studies in Washington, D.C. by Fitzgerald et al. (1982) found no evidence of crystallization or efflorescence when RH was reduced to 30% RH.

Further experimental evidence of the effect of decreasing relative humidity on aerosol size distribution is provided by impactor data reported by Berner (1989) and is shown in Figure 3-31. One impactor sampled aerosol in its humidified state directly from the atmosphere. The inlet of a second impactor was warmed ≈ 7 °C above the ambient temperature of ≈ 5 °C in order to evaporate most of the particle-bound water before collecting the aerosol. The water and other volatile material in both the "wet" and the "dry" samples would evaporate in the laboratory prior to weighing the impactor stages. As can be seen, in the ambient air most of the non-volatile mass was above 1.0 μ m with significant amounts above 2.5 μ m. However, after heating the size of the aerosol was reduced so that most of the non-volatile mass was below 1.0 μ m. Berner treated the distributions as monomodal and derived growth factors of 4.9 for fog and 4.1 for haze. If the observations are treated as multimodal, good bimodal, or as shown in Figure 3-31, trimodal fits are obtained. This splitting into "more" and "less" hygroscopic modes at high relative humidity has been observed by McMurry and co-workers (McMurry and Stolzenberg, 1989; Zhang et al., 1993) (Figure 3-29) and Lowenthal et al. (1995) (Figure 3-30). In some cases, reported by Pitchford and McMurry (1994), splitting into three modes of varying hygroscopicity was observed. However, the separation into two "more" hygroscopic modes may represent, as suggested by Berner, variations in relative humidity extremes during different parts of the overnight sampling period.

In measuring light scattering with the integrating nephelometer, the aerosol community has been very concerned about the difference in relative humidity and temperature in the ambient air and in the volume of air in which particle scattering is actually measured (Covert et al., 1972; Fitzgerald et al., 1982). Temperature differences between the measurement volume and ambient air of 1 or 2 °C can change the relative humidity and change the observed light scattering. Great efforts have been made to minimize this temperature

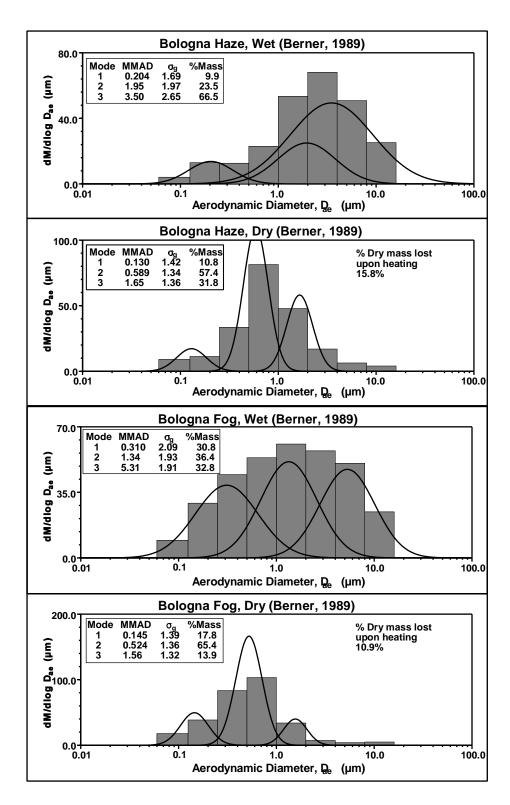


Figure 3-31. Mass size distribution of non-volatile aerosol material. The aerosol was collected at ambient conditions, "wet", or after evaporation of water, "dry".

Source: Berner (1989).

difference. However, researchers have not been nearly as careful in considering temperature and relative humidity effects when measuring size distribution, either with impactors or particle counters, even though effects have been reported in the early literature (Wagman et al., 1967; Sverdrup and Whitby, 1980).

A recent paper by Cass and coworkers (Eldering et al., 1994) provides some insight into how differences in RH resulting from heating can cause differences between particle-counting distributions and impactor distributions. Particle size distributions were obtained by counting particles by mobility (electrical aerosol analyzer) and light scattering (optical particle counter). An example is shown in Figure 3-32. Almost no particles were found between 1.0 and 2.5 µm diameter. When these particle number data were converted to total expected light scattering, they agreed with measurements made by a heated, but not an unheated, integrating nephelometer; and when converted to expected mass, agreed with filter measurements of dry mass. Eldering et al. (1994) conclude that even the moderate heating occurring in mobility and optical counters was enough to change the size of the particles, especially when the ambient air was close to 100% RH. It seems likely that most particle counting systems produce some heating of the aerosol, and thus some reduction of the measured particle size from that existing in the ambient air. On the other hand, if particle-size measuring devices were located in air conditioned or heated trailers or laboratories, the temperature of the sampled air would be changed and the measured particle size distribution would be different from that existing in the ambient air (Sverdrup and Whitby, 1980).

During the high relative humidities that occur at nighttime, growth of hygroscopic components can result in the growth of some fine mode aerosol to diameters greater than 1.0 μ m and perhaps even above 2.5 μ m. As can be seen in Figure 3-28, dry ammonium sulfate particles having a dry diameter of 0.5 μ m will grow to \approx 2.5 μ m at a relative humidity between 99 and 100%. When the relative humidity actually reaches 100%, the particles will continue to grow to maintain the relative humidity at 100%, and eventually become fog droplets that are large enough to be collected in the fraction larger than 2.5 μ m. Ammonium sulfate particles with dry sizes greater than 0.5 μ m would also grow into the larger than 2.5 μ m size range.

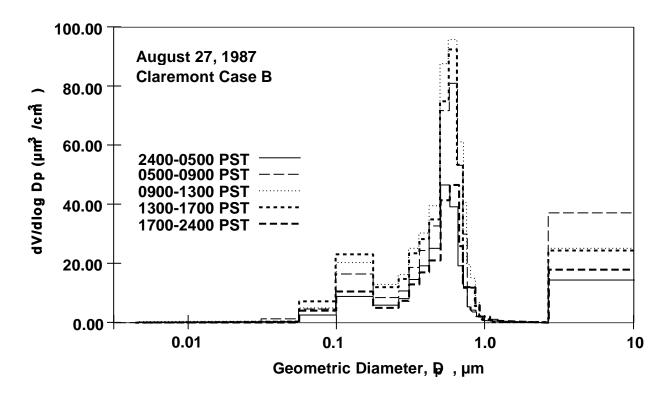


Figure 3-32. Example of particle-counting volume distribution obtained in Claremont, CA. Compare to Figures 3-14 and 3-31. Heating of the sampled air by the mobility and optical counters are believed to have resulted in a distribution representative of a lower than ambient relative humidity.

Source: Eldering et al. (1994).

The addition of water to hygroscopic particles, discussed in the previous section, is a reversible process. Particles absorb water and grow as RH increases; as RH decreases some of the particle-bound water evaporates and the particles shrink. However, the large amount of liquid water associated with hygroscopic particles at high relative humidity provides a medium for liquid phase transformation process. A number of atmospheric process, which convert SO₂ to sulfate or NO_x to nitrate, can take place in water solutions but not in the gas phase. These processes are not reversible but lead to an accumulation of sulfate or nitrate and lead to an increase in the dry size of the particle. Of course as more sulfate or nitrate is added to the particle it will absorb more water so that the wet size will also increase.

The first observation and clear discussion of these combined effects of relative humidity on growth and SO₂ conversion to sulfate are given by Hering and Friedlander (1982) as shown in

Table 3-18. Using a low pressure impactor, they observed that days with higher relative humidity had higher sulfate concentration and higher MMAD's compared to days with lower relative humidity. Hering and Friedlander (1982) named the small mode the condensation mode and suggested that it was formed by the gas phase conversion of SO_2 to sulfate and subsequent nucleation, coagulation, and growth by condensation. They named the larger mode the droplet mode and discussed possible formation mechanisms. This mode is now believed to result from the reaction of SO_2 in fog or cloud droplets (Meng and Seinfeld, 1994).

TABLE 3-18. COMPARISON OF SULFATE CONCENTRATION AND MASS MEAN DIAMETERS OF AEROSOLS FOR DAYS WITH HIGHER AND LOWER RELATIVE HUMIDITY

	Low RH Days	High RH Days
Minimum RH, %	17 - 35	26 - 66
Maximum RH, %	45 - 68	69 - 100
Sulfate concentration, $\mu g/m^3$	3 - 9	3 - 52
Mass median aerodynamic diameter, μ m	0.20 ± 0.02	0.54 ± 0.07

Source: Hering and Friedlander (1982).

In a series of papers McMurry and co-workers make use of the aerosol growth law, originally developed by Heisler and Friedlander (1977), to study the mechanism and rates of sulfate formation in ambient air (McMurry et al., 1981; McMurry and Wilson, 1982, 1983). They were able to apportion growth to condensation and droplet mechanisms and observed droplet growth in particles up to 3 μ m in diameter.

A process of aerosol growth due to increasing relative humidity (Figure 3-33) has also been utilized by Cahill et al. (1990) to explain observations of sulfate size changes during the 1986 Carbonaceous Species Methods Comparison Study in Glendora, CA. Cahill used a DRUM sampler to measure sulfate in nine size ranges. By tracking the mass of sulfate in the 0.56 to 1.15 µm size range Cahill et al. could follow the expansion and contraction of aerosol particles containing sulfate. Because of the relative high time resolution of the DRUM sampler (4 h except for an 8-h increment each night from midnight to 8 a.m.),

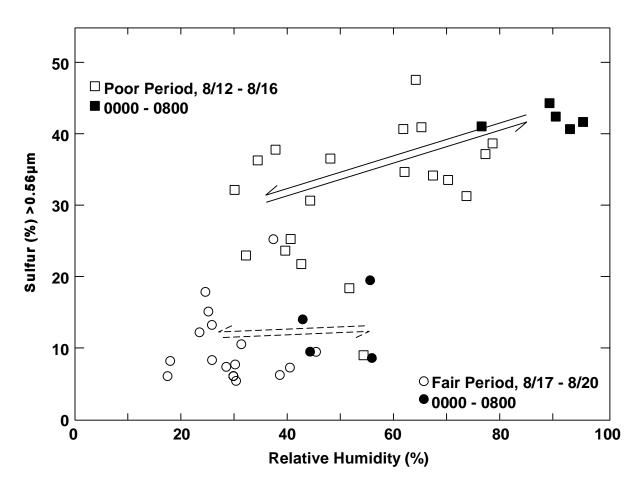


Figure 3-33. Relative humidity versus sulfur, during the 1986 Carbonaceous Species Methods Comparison Study, for particles with $D_{ae}>0.56~\mu m$. The approximate trajectories followed during each day by the $D_{ae}>0.56~\mu m$ sulfur size fraction are shown for period P and period F. Note that even when the humidities are low, 30 to 50 %, the period P aerosols remain coarser by a factor of three than those of period F. The water content incorporated in the aerosols during the 0000- to 0800-h time periods is lost only slowly, giving a strong hysteresis effect in sulfur size.

Source: Cahill et al. (1990).

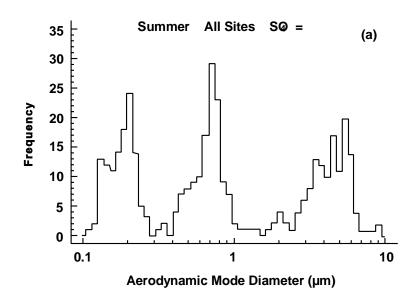
Cahill et al. (1990) could follow this process as the relative humidity increased during the night and decreased during the day. These data indicate that during the "Poor Period" (low visibility) particles grow as relative humidity increases. However, they did not return to the smaller size observed during the "Fair Period" (good visibility). This could be due to a combination of growth due to reaction of SO₂ to sulfate within the particles or failure of the droplet to crystallize thus maintaining particle-bound water in a supersaturated state.

John et al. (1990), in studies in the Los Angeles area, observed a number of sulfate size distributions with MMAD near 1.0 μ m. A histogram of the sulfate MMADs from his study is shown in Figure 3-34. John et al. (1990) have provided a qualitative explanation to account for these large MMADs for fine mode aerosol. In analyzing their data John et al. plotted sulfate mass as a function of sulfate MMAD and found two distinct regions, as shown in Figure 3-35. Distributions with particles near 0.2 μ m diameter are probably still dry; the particles have not reached their deliquescent point. As the relative humidity increases they reach their deliquescent point and grow rapidly into the 0.5 to 0.7 μ m size range. During the formation of fog, the hygroscopic particles act as fog condensation nuclei, and with relative humidity at 100%, grow into 1 to 10 μ m fog droplets. Sulfur dioxide dissolves in the fog droplets and is rapidly oxidized to sulfate by atmospheric oxidants such as H_2O_2 or O_3 , or by catalysis by Fe or Mn. These particles lose some of their water as the relative humidity decreases below 100% RH, but will have substantially more sulfate than prior to activation. Similar processes occur in clouds (Schwartz, 1984a, 1986a).

This type of process probably accounts for the large size of the fine mode observed in Vienna (Berner et al., 1979; Berner and Lürzer, 1980). Winter and summer size distributions are shown in Figure 3-36. Berner et al. reported that fog occurred during the night time during the winter study. In this European study, as in American studies, instances of fine mode size distributions with MMADs near or above 1 µm seem to occur only when fog or very high relative humidity conditions have been present. Two log-normal distributions are fit to the accumulation mode to suggest the separation, at high relative humidity, into hygroscopic and hydrophobic components. No distribution was fit to the coarse mode because only a fraction of the coarse size range was measured.

Similar results have been observed in sampling with dichotomous samplers. A large humidity driven shift of normally fine mode material into the coarse mode was observed by Keeler et al. (1988). In the extreme case, 60% of the $SO_4^=$ and 50% of the $PM_{2.5}$ mass was shifted to the coarse fraction. Such occurrences were not rare, occurring in 12 out of 83 several-hour sampling periods.

In an analysis of data from the IMPROVE network Cahill and co-workers (Eldred et al., 1994) report that 20% of the total sulfate is found in the coarse fraction of PM_{10} . Studies in Philadelphia using dichotomous samplers have also reported that 20% of the total



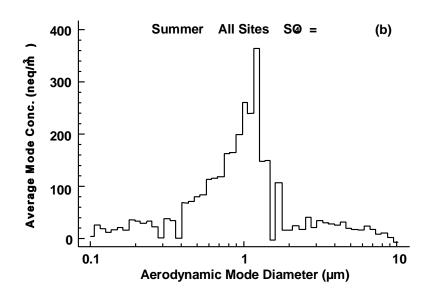


Figure 3-34. Data from the South Coast Air Quality Study (John et al., 1990). Plots show (a) frequency of sulfate modes of various sizes as a function of mode diameter and (b) average sulfate mode concentration as a function of mode diameter. Note that although there are only a few instances when the mode diameter is near 1.0 μ m, it is these situations that give rise to the highest sulfate concentrations. Modes with diameters above 2.5 μ m may be due to collection of fog droplets containing sulfate or reaction of SO₂ in liquid droplets of NaCl due to NaCl sea spray droplets in which SO₂ has dissolved and reacted to form sulfate and release HCl gas.

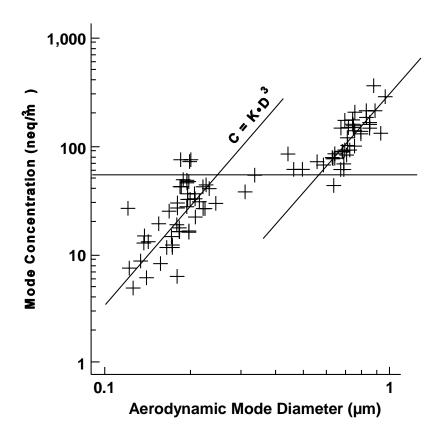


Figure 3-35. Log-log plot of sulfate mode concentration versus aerodynamic mode diameter from Claremont, CA, during the summer SCAQS (John et al., 1990). The solid lines have slopes corresponding to mode concentration increasing with the cube of the mode diameter. A transition between the two modes is believed to occur at approximately the sulfate mode concentration indicated by the horizontal dashed line.

sulfate is found in the coarse fraction (Dzubay et al., 1988). Cahill and coworkers suggest that sulfate particles may grow larger than 2.5 μ m in diameter and thus be sampled in the PM₁₀ fraction but not the PM_{2.5} fraction. It is possible for SO₂ to react with basic carbonate coarse particles to form a sulfate coating or to dissolve in wet NaCl particles, from oceans, lakes, or salt placed on streets to dissolve ice, and be converted to sulfate with the release of HCl. However, there also is substantial evidence that some fine sulfate, and therefore possibly other fine mode material, may be found in the size range above 1.0 μ m and even

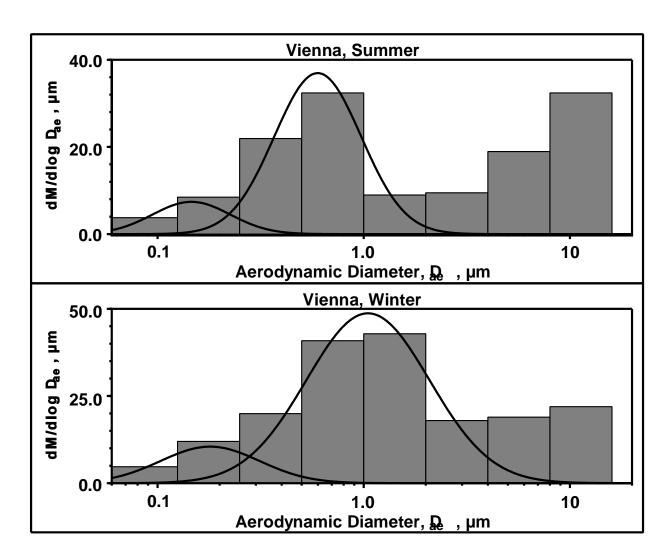


Figure 3-36. Typical results of size-distribution measurements taken with a Berner impactor in a Vienna street with heavy automotive traffic:

(a) measurements taken during summer at three different elevations, (b) measurements taken during winter at three different elevations, fog was frequently present during the winter sampling period.

Source: Berner and Lürzer (1980).

above 2.5 µm diameter, due to the growth of hygroscopic particles at very high relative humidity.

These observations, indicating that, during near 100% relative humidity conditions, significant amounts of normally fine mode material will be found in the coarse fractions (>2.5 μ m diameter), have broader implications than selection of a cut point to separate fine and coarse

particles. Such shifts could cause problems for receptor modeling using chemical mass balance or factor analysis, for interpretation of exposure data in epidemiological studies, and in estimated removal of particulate matter by deposition.

3.7.7 Conclusions

This review of atmospheric particle-size-distributions was undertaken to provide information which could be used to determine what cut-point; $1.0~\mu m$, $2.5~\mu m$, or something in between; would give the best separation between the fine and coarse particle modes. The data do not provide a clear or obvious answer. Depending on conditions, a significant amount of either fine or coarse mode material may be found in the intermodal region between $1.0~and~3~\mu m$. However, the analysis does demonstrate the important role of relative humidity in influencing the size of the fine particle mode and indicates that significant fine mode material is found above $1.0~\mu m$ only during periods of very high relative humidity.

Thus, a PM_{2.5} sample will contain most of the fine mode material, except during periods of RH near 100 %. However, especially in conditions of low RH, it may contain 5 to 20 % of the coarse mode material below 10 μ m in diameter. A PM_{1.0} sample will prevent misclassification of coarse mode material as fine but under high RH conditions will result in some of the fine mode material being misclassified as coarse.

A reduction in RH, either intentionally or inadvertently, will reduce the size of the fine mode. A sufficient reduction in RH will yield a dry fine particle mode with very little material above $1.0~\mu m$. However, reducing the RH by heating will result in loss of semivolatile components such as ammonium nitrate and semivolatile organic compounds. No information was found on techniques designed to remove particle-bound water without loss of other semivolatile components.

3.8 SUMMARY

Atmospheric particulate matter (PM) refers to solid or liquid particles suspended in air. The term atmospheric aerosol refers to both the suspended particles and the air (including gaseous pollutants) in which the particles are suspended. However, the term aerosol is

frequently used to refer only to the suspended particles. The terms particulate matter and particles will be used most frequently in this document.

Particulate matter is not a single pollutant but rather a mixture of many classes of pollutants. The components of PM differ in sources; formation mechanisms; composition; size; and chemical, physical, and biological properties. Particle diameters span more than four orders of magnitude, ranging from a few nanometers (nm) to one hundred micrometers (μ m). Because of this wide size range, plots of particle-size distribution are almost always plotted versus the logarithm of the particle diameter. Diameter usually refers to the aerodynamic diameter, defined as the diameter of a spherical particle with an equal settling velocity but a density of 1 g/cm³. This normalizes particles of different shapes and densities.

One of the most fundamental divisions of atmospheric particles is the naturally occurring separation into a fine particle mode and a coarse particle mode as shown in Figure 3-3. The terms fine mode particles and coarse mode particles are used to refer to particles in the fine or coarse particle distributions. The two distributions overlap between 1 and 3 μ m aerodynamic diameter.

Particles may also be defined by the size cut of the collection or measuring device. A frequently used descriptor is the 50% cut point. This is the aerodynamic diameter at which the efficiency of the device for particle collection is 50%. As particles increase in size above the 50% cut point, they are collected with decreasing efficiency, eventually reaching 0%; as particles decrease in size below the 50% cut point, they are collected with increasing efficiency, eventually reaching 100%. The indicator for the current particle standard is PM_{10} (i.e. particles with a 50% cut point of 10 μ m aerodynamic diameter). However, PM_{10} contains some particles larger than 10 μ m and does not contain all particles below 10 μ m. Fine is also used to refer to particles with an upper cut point of 3.5, 2.5 ($PM_{2.5}$), 2.1, or 1.0 μ m. Coarse is also used to refer to particles between 2.5 and 10 μ m ($PM_{(10-2.5)}$) or particles collected by the high volume sampler as well as the entire coarse mode.

Size fractions may also be characterized in terms of their entrance into various compartments of the body. Thus, inhalable particles enter the respiratory tract, including the head airways. Thoracic particles travel past the larynx and reach the lung airways and the gas-exchange regions of the lung. Respirable particles reach the gas-exchange region of the lung.

 PM_{10} is an indicator of thoracic particles; $PM_{2.5}$ is an indicator of fine mode particles; and $PM_{(10-2.5)}$ is an indicator of the thoracic component of coarse mode particles.

The fine and coarse particle distributions are frequently approximated by log-normal distributions. However, finer distinctions can be made. The fine particles consist of a nuclei mode, composed of particles recently formed from gases, and an accumulation mode, into which the nuclei grow and accumulate (Figure 3-6). Ultrafine particles, defined in this document as distributions with mass median diameters below $0.1~\mu m$, are associated with the nuclei mode (Figures 3-1, 3-2, and 3-13). In the presence of fogs or clouds, the accumulation mode may split into a smaller, less hygroscopic mode and a larger droplet mode. The latter is formed by gases dissolving in the fog or cloud droplets, reacting, and forming particles when the water of the droplets evaporates (Figure 3-14). There may also be several modes within the coarse particle distribution or mode but these are usually less distinct.

The terms primary and secondary, anthropogenic and biogenic, outdoor and indoor microenvironment have significant applications to particulate matter. Primary fine particles are emitted from sources, either directly as particles or as vapors which rapidly condense to form particles. Primary coarse particles are usually formed by mechanical processes. Secondary fine particles are formed within the atmosphere as the result of gas-phase or aqueous-phase chemical reactions. Anthropogenic particles may be formed by primary or secondary processes. Similarly, biogenic particles include primary particles of biological origin, including bioallergens, as well as secondary particles formed from biogenic precursors such as terpenes emitted into the atmosphere. The term outdoor refers to community atmospheres. These are the atmospheres which are usually monitored for particulate matter. Indoor microenviroments include homes, apartments, schools, office buildings and other indoor work places, large enclosed areas such as malls, vehicles used for commuting, etc.

Some general classes of particles, such as organic particles, can occur not only as fine or coarse particles, but can be of either anthropogenic and biogenic origin, and can be produced both in outdoor and indoor microenvironments. Organic particles also can be present in air as primary fine particles from combustion processes or as secondary fine particles formed as a result of atmospheric reactions involving higher molecular weight volatile anthropogenic

alkenes and aromatics or from the atmospheric reactions of volatile biogenic compounds such as terpenes. Therefore, there is considerable overlap for chemical species among the categories listed above.

A substantial fraction of the fine particle mass, especially during the warmer months of the year, is secondary PM, formed as a result of atmospheric reactions. Such reactions involve the gas phase conversion of SO_2 to H_2SO_4 by OH radicals and aqueous-phase reactions of SO_2 with H_2O_2 , O_3 , or O_2 (catalyzed by Fe and Mn). The NO_2 portion of NO_x can be converted to HNO_3 by reaction with OH radicals during the day. During nighttime NO_2 is converted into HNO_3 by a series of reactions involving O_3 and the nitrate radical (NO_3). Both H_2SO_4 and HNO_3 react with atmospheric ammonia (NO_3). Gaseous NO_3 reacts with gaseous O_3 and neutral (O_3). Gaseous O_3 and O_3 reacts with O_3 and O_3 and neutral (O_3). A number of volatile organic compounds can react with O_3 and/or OH radical to form fine organic particles. In addition, acid gases such as O_3 and O_3 may react with coarse particles such as O_3 and O_3

The concentrations of OH radicals, O_3 , and H_2O_2 , formed by gas phase reactions involving volatile organic compounds and NO_x , depend on the concentrations of the reactants, and on meteorological conditions including temperature, solar radiation, wind speed, mixing volume and passage of high pressure systems. Therefore, formation of a substantial fraction of fine particles can depend on the gas phase reactions which also produce O_3 and a variety of other volatile products.

The fine particle fraction, in addition to SO₄⁻ and NO₃⁻, contains elemental carbon (EC), organic carbon (OC), H⁺ (hydrogen ions or acidity) and a number of metal compounds at lower concentrations. Species such as SO₄⁻, NO₃⁻ and some organic species are associated with substantial amounts of particle-bound water. NH₄NO₃ is in equilibrium with HNO₃ and NH₃ so it can vaporize from particles. Organic particles can also be in equilibrium with their vapor. Such species are called semi-volatile. A number of trace elements including, but not necessarily limited to, Pb, Zn, Ni, Cd, Na, Cl, Br, Se and As have been measured in the PM_{2.5} fraction of fine particles. The coarse particles are largely composed of the crustal elements Si, Ca, Al, and Fe. However, a considerable number of elements are found in both the fine and coarse fractions.

Chemical reactions of SO_2 and NO_x within plumes are an important source of H^+ , SO_4 and NO_3^- . These conversions can occur by gas-phase and aqueous-phase mechanisms.

In point-source plumes emitting SO_2 and NO_x , the gas-phase chemistry depends on plume dilution, sunlight and background volatile organic compounds mixed into the diluting plume. For the conversion of SO_2 to H_2SO_4 , the gas-phase rate in such plumes during summer midday conditions in the eastern United States typically varies between 1 and 3% h^{-1} but in the cleaner western United States rarely exceeds 1% h^{-1} . For the conversion of NO_x to HNO_3 , the gas-phase rates appear to be approximately three times faster than the SO_2 conversion rates. Winter rates for SO_2 conversion were approximately an order of magnitude lower than the summer rates.

The contribution of aqueous-phase chemistry to particle formation in point-source plumes is highly variable, depending on the availability of the aqueous phase (wetted aerosols, clouds, fog, and light rain) and the photochemically generated gas-phase oxidizing agents, especially H_2O_2 for SO_2 chemistry. The in-cloud conversion rates of SO_2 to SO_4 can be several times larger than the gas-phase rates given above. Overall, it appears that SO_2 oxidation rates to SO_4 by gas-phase and aqueous-phase mechanisms may be comparable in summer, but aqueous phase chemistry may dominate in winter.

In the western United States, markedly higher SO_2 conversion rates have been reported in smelter plumes than in power plant plumes. The conversion is predominantly by a gas-phase mechanism. This result is attributed to the lack of NO_x in smelter plumes. In power plant plumes NO_2 depletes OH and competes with SO_2 for OH.

In urban plumes, the upper limit for the gas-phase SO_2 conversion rate appears to be about 5% h^{-1} under the more polluted conditions. For NO_2 , the rates appear to be approximately three times faster than the SO_2 conversion rates. Conversion rates of SO_2 and NO_x in background air are comparable to the peak rates in diluted plumes. Neutralization of H_2SO_4 formed by SO_2 conversion increases with plume age and background NH_3 concentration. If the NH_3 concentrations are more than sufficient to neutralize H_2SO_4 to $(NH_4)_2SO_4$, the HNO_3 formed from NO_x conversions may be converted to NH_4NO_3 .

The lifetimes of particles vary with size. Coarse particles can settle rapidly from the atmosphere within hours, and normally travel only short distances. However, when mixed high into the atmosphere as in dust storms the smaller sized coarse mode particles may have longer lives and travel distances. Nuclei mode particles rapidly grow into the accumulation mode. However, the accumulation mode does not grow into the coarse mode. Accumulation-mode fine particles are kept suspended by normal air motions and have very low deposition rates to

surfaces. They can be transported thousands of km and remain in the atmosphere for a number of days. Both accumulation-mode and nuclei-mode (or ultrafine) particles have the ability to penetrate deep into the lungs. Dry deposition rates are expressed in terms of a deposition velocity which varies as the particle size, reaching a minimum between 0.1 and 1.0 μ m aerodynamic diameter. Accumulation-mode particles are removed from the atmosphere primarily by cloud processes. Fine particles, especially particles with a hygroscopic component, grow as the relative humidity increases, serve as cloud condensation nuclei, and grow into cloud droplets. If the cloud droplets grow large enough to form rain, the particles are removed in the rain. Falling rain drops impact coarse particles and remove them. Ultrafine or nuclei mode particles are small enough to diffuse to the falling drop and be removed. Falling rain drops, however, are not effective in removing accumulation-mode particles.

There are many reasons for wanting to collect fine and coarse particles separately. However, because fine-mode particles and coarse-mode particles overlap in the size range between 1.0 and 3 μ m diameter, it is not clear what 50% cut point will give the best separation.

A review of atmospheric particle-size-distribution data did not provide a clear or obvious answer. Depending on conditions, a significant amount of either fine or coarse mode material may be found in the intermodal region between 1.0 and 3 μ m. However, the analysis of the existing data did demonstrate the important role of relative humidity in influencing the size of the fine particle mode and indicated that significant fine mode material is found above 1.0 μ m only during periods of very high relative humidity.

Thus, a PM_{2.5} sample will contain most of the fine mode material, except during periods of RH near 100 %. However, especially in conditions of low RH, it may contain 5 to 20 % of the coarse mode material below 10 μ m in diameter. A PM_{1.0} sample will prevent misclassification of coarse mode material as fine but under high RH conditions will result in some of the fine mode material being misclassified as coarse.

A reduction in RH, either intentionally or inadvertently, will reduce the size of the fine mode. A sufficient reduction in RH will yield a dry fine particle mode with very little material above $1.0~\mu m$. However, techniques to reduce the RH without loss of semivolatile components such as ammonium nitrate and semivolatile organic compounds have not yet been developed.

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